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Heavy metal contamination assessment of surface sediments of the East Zhejiang coastal area during 2012–2015

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ABSTRACT

This work is the first systematic quantitative analysis of the heavy metal situation along the Zhejiang coastal region focusing on the integrative assessment of the concentrations of seven heavy metals (Cu, Cd, Hg, Zn, Pb, Cr, and As) in surface sediments during the 2012–2015 period. Different heavy metal contamination indices were used for surface sediment quality assessment purposes. The numerical results revealed a noticeable spatial fluctuation of the degree of contamination throughout the region during the four years considered. Higher contamination levels and ecological risks were detected in the southern part of the Zhejiang coastal region. It was found that the Cu, Cd and Hg were the predominant contaminants along the Zhejiang coast with mean regional concentrations varying between 29.1 and 34.2, 0.12–0.17, and 0.044–0.052 mg/kg, respectively. The Cr and Pb exhibited lower contamination levels than the other metals during each one of the years 2012–2015. Stochastic site indicators of heavy metal contamination were used to assess regional uncertainties and obtain useful physical interpretations of the state of contamination of the Zhejiang coast. These indicators can be expressed explicitly in terms of probabilities of heavy metal contamination (either at a global scale or spatially distributed over the coastal region), and therefore they can be considered as risk indicators. It was found that the fraction of the coastal region where excess contamination occurred could never exceed the ratio of the mean heavy metal contamination over the selected threshold. In half of the coast study region, the degree of heavy metal contamination was higher than the median spatial contamination values during the month of August of the years 2012–2015. The spatial means of excess contamination and excess differential contamination increased as the relative area of over-contamination increased. Within the substantially contaminated sub-region of the Zhejiang coast, stronger contamination correlations were observed between locations separated by shorter distances. These correlations were higher when smaller thresholds were considered. As regards the spatial connectivity of the corresponding contamination risks, it was found that 44%, 31%, 39% and 63% of the location pairs in the Zhejiang coast simultaneously experienced moderate risks during the years 2012, 2013, 2014 and 2015, respectively. The ratio of the probability of excess contamination at both locations separated by distances < 20 km over the probability of excess contamination at either one of these two locations was high even for large thresholds, indicating that locations with high contamination are concentrated rather than being dispersed along the Zhejiang coast. Lastly, another interesting finding is that the characterization of the Zhejiang coastal region as over-contaminated is very sensitive to the *DC* threshold considered, that is, a small increase in the threshold selected can reduce significantly the probability that region is characterized as over-contaminated.

1. Introduction

Sediments are a vital part of aquatic environments and can be valuable indicators of contaminant monitoring (Harikumar and Nasir, 2010; Suresh et al., 2012; Pejman et al., 2015). Because of their ubiquity, toxicity, persistence, bioaccumulation and biomagnifications, heavy metals accumulated in sediments can be extremely harmful to marine ecosystems and, hence, they have received due attention (Shine et al., 1995; Pan and Wang, 2012; Gui et al., 2017). They can be absorbed by marine organisms and pose a threat to human health through the subsequent bioaccumulation along the food chain (Bryan, 1992, Wang, 2002).

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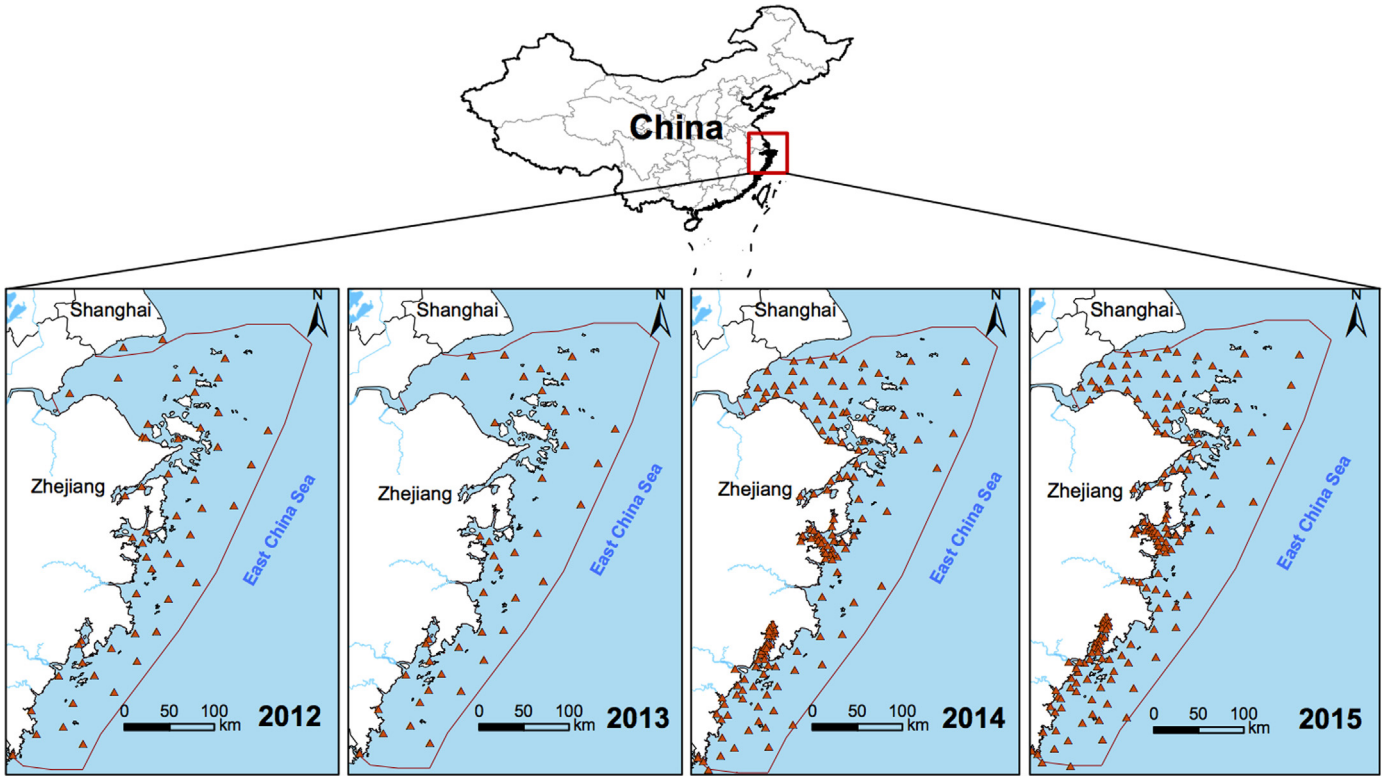

Fig. 1. Study domain (Zhejiang coast along East China Sea), and sampling sites distribution (triangle symbols) during August for the years 2012 through 2015.

Coastal areas at the land-sea interface are vulnerable to environmental changes and human activities. Because of the coastal cities' economic development, industrial expansion and overpopulation, there is an increasing pressure from pollutant loadings on coastal ecosystem. A considerable amount of heavy metals is deposited into the coastal and estuarine environments due to runoff and anthropogenic input (industrial waste, domestic sewage, ship trash etc). They can heavily accumulate in marine sediments, which act as a sink for these contaminants. Moreover, under certain conditions created by natural or anthropogenic impacts (geological disturbance, earthquake, salinity or pH change), the contaminants may be released back to water columns causing secondary contamination (Shipley et al., 2011, Kalnejais et al., 2010). Hence, sediment analysis can be an important component of a heavy metal contamination study in coastal regions that provide numerous ecosystem services (i.e., scientific research, sea food, biodiversity) to humans.

During the last decades, several studies have been published of marine sediment heavy metal contamination for different coastal areas (Zhang et al., 2009; Gao and Chen, 2012; Chen et al., 2016; Lu et al., 2017). In these studies, various indices (Contamination factor-CF, Degree of contamination-DC, Enrichment factor-EF, and Index of geoaccumulation-I_{geo}) were applied to assess the degree of contamination and ecological risk (see SI Table S1). These indices were established by comparing contamination results *vs*. background values and sediment quality guidelines, or by using aggregation methods to assess the degree of contamination (Caeiro et al., 2005; Vm et al., 2016). It should be noticed that most of these studies were based on in-suit site-specific analysis, which involves considerable uncertainty.

Based on the comprehensive monitoring of seven heavy metals (copper, Cu, cadmium, Cd, mercury, Hg, zinc, Zn, lead, Pb, chromium, Cr, and arsenic, As), the main goal of the present work is to investigate the evolution of these heavy metal contamination of Zhejiang's near sea region (which has not been studied before). A set of standard environmental indices together with a set of stochastic site indicators developed by the authors were used in this work. In particular, this work involved three main stages: (1) Explore the spatial distributions and physical correlations of the heavy metals; (2) assess the contamination level and potential risk based on standard global indices (contamination factor, geo-accumulation index, and ecological risk index); and (3) quantify the contamination uncertainty and regional variability using a set of stochastic site indicators (local and global). The results of this study could provide a comprehensive assessment of heavy metal contamination in the sediments of the Zhejiang coastal region. They can also aid the development of effective remediation strategies and improve government decision-making and coastal management.

2. Materials and methods

2.1. Study area and data processing

The study domain, with an area of about 44.4 thousand km^2, is located in the eastern part of Zhejiang province, China (27.0°-31.0°N, 120.4°-123.5°E), which belongs to the East China Sea. During the last decades, Zhejiang province has experienced rapid economic development and the near sea region is characterization by high population density, and developed shipping industry and aquaculture. Every year, large amounts of pollutants are transferred into rivers (Qiantang river, Yong river, Ou river etc) through rain wash and underground water immigration and then discharged into the ocean. The heavy metals produced by mining plants, e-waste processing sites or domestic sewage are subsequently deposited on marine sediments. Yet, very few studies have investigated the heavy metal contamination at the particular regional scale (Chen et al., 2016; Yang et al., 2017). The Zhoushan fishing ground, one of the biggest fishery in China, is located in the north of the Zhejiang coastal waters. A large amount of seafood is produced each year, which is consumed by local people or exported to other provinces. Thus, the comprehensive monitoring of heavy metal contamination in this area is essential for coastal environment management and the rigorous assessment of ecosystem and human exposure to heavy metals.